A low-dimension structure strategy for flexible photodetectors based on perovskite nanosheets/ZnO nanowires with broadband photoresponse

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ABSTRACT Flexible photodetectors (PDs) have huge potential for application in next-generation optoelectronic devices due to their lightweight design, portability, and excellent large area compatibility. The main challenge in the construction of flexible PDs is to maintain the optoelectronic performance during repetitive bending, folding and stretching. Herein, flexible PDs based on ZnO nanowires (NWs) and CsPbBr₃ nanosheets (NSs) were constructed by an integrated low-dimensional structure strategy. Benefiting from the flexibility of unique sheet and wire structures, the PDs were able to maintain excellent operational stability under various mechanical stresses. For example, the PDs exhibited no obvious changes in optoelectronic performance after bending for 1000 times. Additionally, the PDs exhibited an integrated broadband response ranging from ultraviolet to visible region due to the combination of the intrinsic light absorption capability of ZnO and CsPbBr₃. The PDs demonstrated high responsivities of 3.10 and 0.97 A W⁻¹ and detectivities of 5.57×10¹² and 1.71×10¹² Jones under ultraviolet and visible light irradiation, respectively. The proposed construction strategy for highly flexible and performance-integrated PDs shows great potential in future smart, wearable optoelectronic devices.

Keywords: perovskite, ZnO nanowires, flexible photodetectors, broad spectrum

INTRODUCTION

Flexible electronic devices with highly integrated, compact, and portable features are very attractive in the fields of smart photodetectors (PDs) [1–7], wearable sensors [8–11], bendable solar cells [12–15], and field-emission transistors [16–19]. Among various flexible devices, PDs have aroused extensive attention due to their wide ap-

plications in flame detection, optical communication, and environmental monitoring [20–25]. In general, flexible PDs are fabricated on bendable or ductile substrates (such as plastic, metal sheets, and rubber), which requires the selected semiconductor materials possessing the ability to operate well while bent, rolled or stretched. Thus, developing suitable semiconductor materials for flexible PDs is highly desirable.

Generally, thin-film materials and low-dimensional materials are employed for the construction of flexible PDs. However, the optoelectronic performance of thin film-based devices may deteriorate because of the structural damage caused by sustained stretching stress. In addition, cracks can easily extend from the top to the bottom of the film under mechanical stress, which will block the carrier transport and eventually lead to degradation, even the expiration of the PDs [26–28]. Whereas, cracking in low-dimensional materials can be prevented because the generation potential of crack decreases linearly with decreasing dimensions. Furthermore, materials with low-dimensional nanostructures have much higher photo-sensitivity and faster carrier transport. Consequently, low-dimensional materials with excellent optical and electrical properties possess broad prospect in flexible PDs.

Among the various low-dimensional materials, the zero dimensional (0D) nanoparticle-based film is similar to bulk film, which also suffers from cracks under repeated mechanical stresses. Therefore, 0D nanoparticles are not suitable for flexible PDs. The distinct geometric structures of one-dimensional (1D) nanowires (NWs) and two-dimensional (2D) nanosheets (NSs) make them reliable and suitable for flexible PDs [29–31]. For 1D NWs, they

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intertwine with each other to form a film of dense network that could resist external forces. Meanwhile, outstanding electrical properties of 1D NWs facilitate efficient carrier transport. The effective area of the carriers is limited and the transit time is shortened due to the reduced dimensionality, which enhances the photo-response and optical gain [32–36]. Moreover, NW-based films can be easily transferred to flexible substrates. As a result, NW-based PDs have high performances and broad application prospects in the flexible fields. For example, Kind *et al.* [37] reported an ultraviolet PD based on an individual ZnO NW and demonstrated its potential for optical switches. Wang *et al.* [38] described a ZnGa_2O_4 PD with a large photoresponse resulting from the Schottky barrier between the Ag and ZnGa_2O_4 NWs and the junction barrier between interconnecting ZnGa_2O_4 NWs. Li *et al.* [39] demonstrated that PDs based on ZnO quantum dot-decorated Zn_2SnO_4 NWs exhibited the improved responsivity and outstanding flexibility. With respect to 2D materials, such as NSs, they would interconnect with each other in films, exhibiting superior flexibility and compatibility than quantum dots and bulk materials. Another advantage of 2D materials is that charges can be transported rapidly in plane due to smaller hopping barriers [40–41]. As a consequence, 2D materials display distinctive superiority in applications of flexible PDs. For example, Song *et al.* [42] reported 2D inorganic perovskite NSs for the first time and fabricated the PD with a high and fast photoresponse. Lim *et al.* [43] investigated a flexible PD based on a homogeneous MoS_2 layer, which decreased only 5.6% in the photocurrent after bending for 10^5 cycles. Thus, 2D NSs have also been proved as promising materials for flexible PDs.

With the rapid development of society and technology, it is difficult to satisfy the large demand for multifunctional detection by using a single material. Therefore, integrating different nanomaterials is a feasible strategy for constructing multifunctional devices. Herein, a highly flexible PD based on low-dimensional ZnO NWs and CsPbBr_3 NSs is reported for the first time. The PD presents a low dark current, a high on/off ratio ($>10^3$), good mechanical durability, and superior flexibility (>1000 cycles). Because ZnO is a typical semiconductor material with a bandgap of 3.3 eV and absorption in the ultraviolet region, and CsPbBr_3 is considered as an emerging material with excellent absorption in the visible region. Thus, the PD demonstrates broad photodetecting performance resulting from the integration of ZnO and CsPbBr_3 . The fabricated PD responses to light ranging from ultraviolet to visible region and presents high detectivities up to

5.57×10^{12} and 1.71×10^{12} Jones at the wavelengths of 380 and 520 nm, respectively.

EXPERIMENTAL SECTION

Preparation of CsPbBr_3 NSs

The CsPbBr_3 NSs were synthesized *via* a solution process at room temperature. First, 0.1064 g CsBr and 0.0918 g PbBr_2 were dissolved in 7 mL dimethyl sulfoxide (DMSO) and 0.5 mL acetic acid as solution A. 1 g octadecylamine was dissolved in 5 mL acetic acid using an ultrasonic treatment for a few minutes as solution B. Then 200 μL of solution A and 1 mL of solution B were mixed in a 50-mL centrifuge tube and stirred for 2 min. Later, 20 mL toluene was added quickly into the tube. The mixture was stirred for another 3 min to form orange CsPbBr_3 NSs. Finally, pure CsPbBr_3 NSs were obtained by rinsing with toluene and centrifugation.

Fabrication of the flexible PDs

A polyethylene terephthalate (PET) substrate with Au/Cr interdigital electrodes was used for constructing PDs. Firstly, 0.05 g ZnO NWs (purchased from Hebei ShaoJing New Material Technology Co., Ltd.) were dispersed in 20 mL isopropanol and CsPbBr_3 NSs were dispersed in 4 mL toluene. Then ZnO NWs were deposited onto a polyvinylidene fluoride (PVDF) filtration membrane by suction filtration. Finally, the filtration membrane with the ZnO NWs and CsPbBr_3 NSs was overturned and quickly transferred to the aforementioned PET/Au substrate by applying appropriate pressure. When the filtration membrane was uncovered, the ZnO/ CsPbBr_3 film was laid onto the PET substrate.

Characterization of materials and devices

The morphologies of the NWs and NSs were observed by field emission scanning electron microscopy (FESEM, FEI Quanta 250F) and transmission electron microscopy (TEM, FEI Tecnai G20). The X-ray diffraction (XRD) patterns were obtained by using Bruker D8 Advance. The thickness of the CsPbBr_3 NS was measured by a Bruker Multimode 8. The absorbance of the homogeneous dispersion was measured by using a Shimadzu 3600 UV-Vis spectrophotometer. The responsivity and external quantum efficiency (EQE) of PDs were measured through a Zolix DSR-101-UV detector responsivity measurement system. 320 and 532 nm continuous semiconductor lasers were used as light source to measure the optoelectronic properties of the PDs. A Keithley 6487 picoammeter was used to obtain the current.

RESULTS AND DISCUSSION

Morphologies and structures

The crystal structures of the ZnO NWs and ultrathin CsPbBr₃ NSs were analyzed by TEM and XRD. As shown in Fig. 1a and Fig. S1, the NW has a small diameter of approximately 100 nm and a large length to diameter ratio. The structural information was obtained by selected-area electron diffraction (SAED), as shown in the inset of Fig. 1a. The lattice spacing of 0.26 nm corresponds to the crystal plane (001) of ZnO, and the ZnO NWs have a preferred growth orientation along the (002) direction. Fig. 1b shows that the 2D quadrilateral CsPbBr₃ NSs have clean and smooth surfaces. No other by-pro-

ducts are observed, such as small-sized particles, which are common in the synthesis of CsPbBr₃ nanocrystals [44]. The NSs have similar length and width of several hundred nanometers, and the thickness is approximately 8 nm, as shown in the atomic force microscopy (AFM) image (Fig. S2). Furthermore, the NS in the bottom can be observed and the number of stacked layers of NSs can be determined. In addition, the wrinkles on the NSs are clear evidence of good flexibility. The insets in Fig. 1b reveal that the lattice spacing is 0.58 nm, corresponding to the (100) plane, which is in agreement with the XRD patterns presented in Fig. 1c. In the XRD patterns of the ZnO NWs and the ZnO/CsPbBr₃ assembled film, no impurity peaks are observed, demonstrating superior

Figure 1 Typical TEM images of (a) ZnO NW and (b) CsPbBr₃ NSs, insets are the corresponding (i) HRTEM and (ii) SAED. (c) XRD patterns of the pure ZnO NWs and CsPbBr₃/ZnO heterojunction film. Absorbance and PL spectra of (d) ZnO NWs and (e) CsPbBr₃ NSs.

purity of the ZnO NWs and the CsPbBr₃ NSs. The characteristic peaks of (100), (110), and (200) observed in both patterns are indexed to the wurtzite structure of ZnO. A sharp (100) diffraction peak and two additional relatively small peaks indicate the typical orientation growth of ZnO NWs. Compared with the XRD pattern of the ZnO NWs, two additional peaks of (100) and (200) for CsPbBr₃ (JSPDS #18-0364) appeared. The XRD results indicate that the prepared CsPbBr₃ NSs exhibit the typical layered structure, which is favorable for carrier transport in PDs.

Optical properties

The optical properties of the NWs and NSs were measured by using UV-Vis absorption and photoluminescence (PL) spectra. As seen in Fig. 1d, there is almost no absorption in the visible region, whereas the absorption sharply increases in the ultraviolet region. The exciton peak at 360 nm is attributed to the near-bandgap absorption of the ZnO NWs. The normalized PL spectrum of the ZnO NWs consists of two emission peaks, a near-band-edge (NBE) emission peak centered at 377 nm with a full width at half-maximum (FWHM) of 10 nm, and a broad peak located in the visible region. Usually, the NBE emission is attributed to the collision between excitons, indicating the emergence and recombination of photo-generated carriers in the ZnO NWs. The wide peak around 530 nm is attributed to the deep-level emission from oxygen vacancies on the surface of the ZnO NWs [45]. The oxygen vacancies may absorb oxygen under the atmospheric environment [46–48], which improves the device performance to some extent. As shown in Fig. 1e, the prepared CsPbBr₃ NSs exhibit an absorbance edge at approximately 520 nm, which is consistent with the previous results [49,50]. The corresponding PL spectrum shows a highly symmetric emission peak at 520 nm. Moreover, the emission peak has a

narrow linewidth of 22 nm and no additional sub-band-gap emission is observed, indicating the high quality of the CsPbBr₃ NSs, which is favorable for constructing high-performance devices.

Device construction

The flexible PD was constructed by combining ZnO NWs and CsPbBr₃ NSs (Fig. 2). First, the ZnO NWs were deposited on the PVDF filter membrane by a vacuum-filtration route. The corresponding SEM image shows that the highly dense NWs possess a very long length beyond tens of microns and are naturally intertwined with each other. Next, the CsPbBr₃ NSs were deposited on the existing ZnO NWs by vacuum-filtration route. As shown in the SEM image, the NSs deposited on the ZnO NWs are spread out and interlinked. Finally, the assembled film combined with the ZnO NWs and the CsPbBr₃ NSs was transferred onto the PET substrate with Au/Cr interdigital electrodes. The SEM image of the fabricated device demonstrates the morphology of the device with copious ZnO NWs. The Au electrodes can be clearly observed. However, the CsPbBr₃ NSs at the bottom are not clear due to the overlapping ZnO NWs. This transfer method can be used for assembling compact film on various substrates.

The flexibility of the device

The flexibility and bendability of the fabricated PD were then evaluated. Fig. 3a presents a typical photograph of the flexible ZnO/CsPbBr₃ PD. To assess the reliability of the fabricated devices, we compared the device performances before and after repeated mechanical bending. The SEM images of the device before and after bending 1000 times are shown in Fig. 3b and c, where no apparent change is observed, indicating the excellent bendability of the PDs. In addition, the photocurrents under periodical illumination (Fig. 3d) also exhibit no significant at-

Figure 2 Schematic diagrams of the fabrication process of the PD and the corresponding morphologies of the sample at each step.

Figure 3 The bending performance of the flexible PDs. (a) The photograph of the flexible PD. SEM images of the flexible PD (b) before and (c) after bending. (d) I - t curves at 1 V of the flexible PD under flat and bending state. (e) The photoresponse of the flexible PD under different bending times.

tenuation in the bent state compared to the flat state, demonstrating excellent stability and reversibility. The slight change in the photocurrent may be the result of the contacts between the electrodes and probes when the device changes from the flat state to the bent state. Moreover, the photocurrent of the flexible PD under 1 mW illumination after bending for 1000 times (Fig. 3e) is almost the same as that of the initial device. This further demonstrates that the device is adaptable to the bending stress. The behavior of the flexible PD at different bending degrees was also evaluated (Fig. S3). The photographs in the inset show the bending degree of the device. At different bending degrees, the dark currents show no change while the photocurrents show a slight fluctuation. These observations indicate that the fabricated flexible PD based on the ZnO/CsPbBr₃ heterojunction is able to bear an external mechanical force, which can be attributed to the flexibility of the unique low-dimensional nanostructures.

Broadband response of the device

Due to the advantages of the integration of ZnO and CsPbBr₃, the PD can potentially exhibit a broad photo-

response. Fig. 4a shows the absorption of the ZnO/CsPbBr₃ composite, which has a wider range than the ZnO NWs including the visible region due to the introduction of the CsPbBr₃ NSs. Compared with ZnO NWs and CsPbBr₃ NSs, the PL spectrum of ZnO/CsPbBr₃ (Fig. S4a) exhibits no obvious shift. However, we find that the PL lifetime of the ZnO/CsPbBr₃ is shorter than that of pure CsPbBr₃, which is attributed to the transfer of the carriers from CsPbBr₃ to ZnO. Thus, the carriers can be easily separated and transported to the electrode, which would improve the photoelectric properties of the flexible PD (Fig. S4b). The unique optical properties of the ZnO/CsPbBr₃-assembled film make it an ideal candidate for broad-spectrum PDs.

The responsivity (R) is one of the most crucial parameters for evaluating the photoresponse of a PD. R is defined in the following equation [51],

$$R = \frac{I_{\text{photo}}}{P_{\text{light}} \times A}, \quad (1)$$

where I_{photo} , P_{light} and A are the photocurrent, irradiation power density and working area of the PD, respectively. The responsivities were measured under different bias

Figure 4 The broad spectral characteristics of the flexible PD. (a) Absorbance of the ZnO/CsPbBr₃ composite. (b) Responsivity spectrum of ZnO/CsPbBr₃ heterojunction PD under different applied bias. (c) Detectivity of the PD under 9 V.

voltages using a standard Si PD as a reference, as shown in Fig. 4b. The broad-responsivity spectra of the flexible PD cover a wavelength range from 300 nm (ultraviolet region) to 550 nm (visible region), which is consistent with the UV-Vis absorption spectrum. Normally, the peak located at 380 nm is attributed to the ZnO NWs (shown in Fig. S5a) and the 520 nm peak is attributed to the CsPbBr₃ NSs (shown in Fig. S5b). The sharp increases below 380 and 520 nm are owing to the different concentrations of electron-hole pairs excited by various wavelengths of light. The photoconduction mechanism of the PD is based on the production of photo-generated carriers (electron-hole pairs), which is motivated by incident light with greater photon energy than the bandgap. Meanwhile, the responsivity increases with increasing bias voltage, reaching 3.1 A W⁻¹ at 380 nm and 0.97 A W⁻¹ at 520 nm under 9 V, which are higher than those of the reference Si PD (<0.2 A W⁻¹). The EQE (shown in Fig. S6) exhibits a high numerical value of 1024% at 380 nm and 230% at 520 nm under 9 V. The high EQE value is attributed to a large number of photo-generated carriers under irradiation and the additional charges from the applied voltage.

Assuming that the short noise from the dark current acts as the major noise, the detectivity (D^*) [52] is calculated as:

$$D^* = \frac{R \times A^{1/2}}{(2e \cdot I_{\text{dark}})^{1/2}} \quad (2)$$

where e refers to the electron charge. The relationship between the calculated detectivity and the wavelength is shown in Fig. 4c. The detectivities at 380 and 520 nm are 5.57×10^{12} and 1.71×10^{12} Jones, respectively. This is comparable to the reported state-of-the-art PDs with broad-band photoresponse [53–56].

Mechanism of charge transport

The current-voltage (I - V) characteristics of the ZnO/CsPbBr₃ heterojunction PD were measured in dark and light conditions (the power density of the applied laser was 1 mW cm⁻²), respectively. As shown in Fig. 5a, the photocurrents under illumination exhibit a large increase compared with the dark current and an excellent photoresponse is observed in the UV-vis region. The extracted dark current from the I - V logarithmic curve (Fig. 5b) is approximately 5.9×10^{-10} A under an applied

Figure 5 (a) *I-V* characteristics and (b) *I-V* logarithmic characteristics of the flexible ZnO/CsPbBr₃ heterojunction PD under dark and illumination.

voltage of 2 V, while the photocurrents under irradiation of 320 and 532 nm are 2.9×10^{-5} and 5.2×10^{-6} A, respectively. The on/off ratios, which are defined as $(I_{\text{photo}} - I_{\text{dark}})/I_{\text{dark}}$, are 4.91×10^4 and 8.81×10^3 , respectively. These high on/off ratios are the result of the high light absorbance of the ZnO NWs in the ultraviolet region and the CsPbBr₃ NSs in the visible region. The perovskite/ZnO heterojunction PDs have been reported and some of them have demonstrated excellent responsivity, detectivity, and high on/off ratios, as listed in Table 1. These high-performance PDs based on perovskite/ZnO were usually realized on rigid substrates. However, flexible PDs with high optoelectronic properties are relatively scarce on account of substrate compatibility and mechanical stability. In this study, both excellent flexibility and high performance are achieved by using low-dimensional structures.

In order to illustrate the underlying mechanism of the broad photoresponse of the flexible PD, schematic diagrams of the carrier transport are presented in Fig. 6. The band structures of the PD are shown in Fig. 6a and c, demonstrating that a staggered heterojunction of ZnO/CsPbBr₃ is formed. Fig. 6b and d show the transport paths of the electron and hole in the PD under illumination. Under illumination of 320 nm, photo-generated carriers are mainly generated in the ZnO NWs. Because most of

the CsPbBr₃ NSs are covered by the ZnO NWs and hence the light absorbance mainly occurs in the ZnO NWs. Relatively, the valence band of the ZnO lies lower than that of the CsPbBr₃, the photo-generated holes tend to migrate to the CsPbBr₃ NSs, resulting in efficient separation of the photo-generated carriers. Then, carriers pass through the NSs, and are collected by the Au electrode. Under illumination of 532 nm, the long wavelength light normally has a deeper penetration ability. The light can pass through the ZnO NWs and reach the NSs. Because the photon energy is much lower than the bandgap of ZnO, which cannot produce photo-generated carriers in ZnO. Thus, the photo-generated carriers are mainly generated in the CsPbBr₃ NSs and are collected by the electrodes. The different photo-generated carrier features of ZnO NW/CsPbBr₃ NS structure make the flexible PD with a broad photoresponse.

CONCLUSION

In summary, a mechanically stable and flexible PD based on ZnO NWs/CsPbBr₃ NSs was designed by using a simple vacuum-filtration route. The optoelectronic properties of PD exhibited almost no change after bending for 1000 times. Importantly, as a result of the different light absorbance characteristics of ZnO and

Table 1 Performance comparisons of perovskite/ZnO nanostructure PDs

Materials	Substrates	Wavelength (nm)	Responsivity (A W^{-1})	Detectivity (Jones)	On/off ratio	Ref.
CH ₃ NH ₃ PbI ₃ /ZnO nanorod array	FTO glass	500	24.3	3.56×10^{12}	–	[57]
CsPbBr ₃ /ZnO	PET	450	4.25		$>10^4$	[58]
ZnO nanofiber/perovskite	ITO glass		0.1	10^{13}	2.80×10^3	[59]
CH ₃ NH ₃ PbI ₃ /ZnO	Mica	380	4	1.74×10^9	8	[60]
CsPbBr ₃ NSs/ZnO NWs	PET	380	3.1	5.57×10^{12}	4.91×10^4	This work
		520	0.97	1.71×10^{12}	8.81×10^3

Figure 6 The energy band diagram in ZnO/CsPbBr₃ heterojunction under illumination of (a) 320 nm and (c) 532 nm, respectively. The schematic diagram of the carrier transport in ZnO/CsPbBr₃ heterojunction under illumination of (b) 320 nm and (d) 532 nm, respectively.

CsPbBr₃, the flexible PD exhibited a broadband photo-response. High responsivities of 3.10 and 0.97 A W⁻¹ and high detectivities of 5.57×10¹² and 1.71×10¹² Jones were obtained in the ultraviolet and visible regions, respectively. These results demonstrate that the low-dimensional materials show great advantages in flexible optoelectronic devices, which makes them a favorable candidate in the fields of wearable electronic devices, folding displays and other applications.

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- Hu PA, Wang L, Yoon M, *et al.* Highly responsive ultrathin gas nanosheet photodetectors on rigid and flexible substrates. *Nano Lett*, 2013, 13: 1649–1654
- Tian W, Zhang C, Zhai T, *et al.* Flexible ultraviolet photodetectors with broad photoresponse based on branched ZnS–ZnO heterostructure nanofilms. *Adv Mater*, 2014, 26: 3088–3093
- Zhou X, Zhang Q, Gan L, *et al.* Large-Size growth of ultrathin SnS₂ nanosheets and high performance for phototransistors. *Adv Funct Mater*, 2016, 26: 4405–4413
- Wang S, Zou Y, Shan Q, *et al.* Nanowire network-based photodetectors with imaging performance for omnidirectional photo-detecting through a wire-shaped structure. *RSC Adv*, 2018, 8: 33666–33673
- Lou Z, Li L, Shen G. High-performance rigid and flexible ultra-violet photodetectors with single-crystalline ZnGa₂O₄ nanowires. *Nano Res*, 2015, 8: 2162–2169
- Xie C, Yan F. Flexible photodetectors based on novel functional materials. *Small*, 2017, 13: 1701822
- Wang X, Song W, Liu B, *et al.* High-performance organic-inorganic hybrid photodetectors based on P3HT:CdSe nanowire heterojunctions on rigid and flexible substrates. *Adv Funct Mater*, 2013, 23: 1202–1209
- Choi S, Lee H, Ghaffari R, *et al.* Recent advances in flexible and stretchable bio-electronic devices integrated with nanomaterials. *Adv Mater*, 2016, 28: 4203–4218
- Xue J, Chen J, Song J, *et al.* Wearable and visual pressure sensors based on Zn₂GeO₄@polypyrrole nanowire aerogels. *J Mater Chem C*, 2017, 5: 11018–11024
- Gao W, Emaminejad S, Nyein HYY, *et al.* Fully integrated wearable sensor arrays for multiplexed *in situ* perspiration analysis. *Nature*, 2016, 529: 509–514
- Amjadi M, Pichitpajongkit A, Lee S, *et al.* Highly stretchable and sensitive strain sensor based on silver nanowire-elastomer nanocomposite. *ACS Nano*, 2014, 8: 5154–5163
- Shin SS, Yang WS, Noh JH, *et al.* High-performance flexible perovskite solar cells exploiting Zn₂SnO₄ prepared in solution below 100°C. *Nat Commun*, 2015, 6: 7410
- Kaltenbrunner M, Adam G, Glowacki ED, *et al.* Flexible high power-per-weight perovskite solar cells with chromium oxide-metal contacts for improved stability in air. *Nat Mater*, 2015, 14: 1032–1039
- Wang Y, Bai S, Cheng L, *et al.* High-efficiency flexible solar cells based on organometal halide perovskites. *Adv Mater*, 2016, 28:

- 4532–4540
- 15 Kim T, Kim JH, Kang TE, *et al.* Flexible, highly efficient all-polymer solar cells. *Nat Commun*, 2015, 6: 8547
 - 16 Zang Y, Zhang F, Huang D, *et al.* Flexible suspended gate organic thin-film transistors for ultra-sensitive pressure detection. *Nat Commun*, 2015, 6: 6269
 - 17 Yang Y, Yang X, Zou X, *et al.* Ultrafine graphene nanomesh with large on/off ratio for high-performance flexible biosensors. *Adv Funct Mater*, 2017, 27: 1604096
 - 18 Xiang L, Wang Z, Liu Z, *et al.* Inkjet printed flexible biosensor based on graphene field effect transistor. *IEEE Sens J*, 2016, 16: 8359
 - 19 Petrone N, Chari T, Meric I, *et al.* Flexible graphene field-effect transistors encapsulated in hexagonal boron nitride. *ACS Nano*, 2015, 9: 8953–8959
 - 20 Lee SH, Kim SB, Moon YJ, *et al.* High-responsivity deep-ultraviolet-selective photodetectors using ultrathin gallium oxide films. *ACS Photonics*, 2017, 4: 2937–2943
 - 21 Jayasinghe RC, Ariyawansa G, Dietz N, *et al.* Simultaneous detection of ultraviolet and infrared radiation in a single GaN/GaAlN heterojunction. *Opt Lett*, 2008, 33: 2422–2424
 - 22 Monroy E, Omnis F, Calle F. Wide-bandgap semiconductor ultraviolet photodetectors. *Semicond Sci Technol*, 2003, 18: R33–R51
 - 23 Liu K, Sakurai M, Aono M. ZnO-based ultraviolet photodetectors. *Sensors*, 2010, 10: 8604–8634
 - 24 Su L, Yang W, Cai J, *et al.* Self-powered ultraviolet photodetectors driven by built-in electric field. *Small*, 2017, 13: 1701687
 - 25 Dong Y, Wang S, Zou Y, *et al.* Zinc stannate nanocrystal-based ultrarapid-response UV photodetectors. *Adv Mater Technol*, 2018, 3: 1800085
 - 26 Cheng W, Niederberger M. Evaporation-induced self-assembly of ultrathin tungsten oxide nanowires over a large scale for ultraviolet photodetector. *Langmuir*, 2016, 32: 2474–2481
 - 27 Li X, Yu D, Cao F, *et al.* Healing all-inorganic perovskite films via recyclable dissolution-recrystallization for compact and smooth carrier channels of optoelectronic devices with high stability. *Adv Funct Mater*, 2016, 26: 5903–5912
 - 28 Sang L, Liao M, Sumiya M. A comprehensive review of semiconductor ultraviolet photodetectors: From thin film to one-dimensional nanostructures. *Sensors*, 2013, 13: 10482–10518
 - 29 Liu Z, Xu J, Chen D, *et al.* Flexible electronics based on inorganic nanowires. *Chem Soc Rev*, 2015, 44: 161–192
 - 30 Yan R, Gargas D, Yang P. Nanowire photonics. *Nat Photon*, 2009, 3: 569–576
 - 31 Li Y, Qian F, Xiang J, *et al.* Nanowire electronic and optoelectronic devices. *Mater Today*, 2006, 9: 18–27
 - 32 Peng M, Wen Z, Shao M, *et al.* One-dimensional CdS_xSe_{1-x} nanoribbons for high-performance rigid and flexible photodetectors. *J Mater Chem C*, 2017, 5: 7521–7526
 - 33 Shahid M, Cheng J, Li T, *et al.* High photodetectivity of low-voltage flexible photodetectors assembled with hybrid aligned nanowire arrays. *J Mater Chem C*, 2018, 6: 6510–6519
 - 34 Um DS, Lee Y, Lim S, *et al.* High-performance MoS₂/CuO nanosheet-on-one-dimensional heterojunction photodetectors. *ACS Appl Mater Interfaces*, 2016, 8: 33955–33962
 - 35 Liang FX, Wang JZ, Li ZP, *et al.* Near-infrared-light photodetectors based on one-dimensional inorganic semiconductor nanostructures. *Adv Opt Mater*, 2017, 5: 1700081
 - 36 Yang X, Liu Y, Lei H, *et al.* An organic–inorganic broadband photodetector based on a single polyaniline nanowire doped with quantum dots. *Nanoscale*, 2016, 8: 15529–15537
 - 37 Kind H, Yan H, Messer B, *et al.* Nanowire ultraviolet photodetectors and optical switches. *Adv Mater*, 2002, 14: 158–160
 - 38 Wang J, Yan C, Lin MF, *et al.* Solution-assembled nanowires for high performance flexible and transparent solar-blind photodetectors. *J Mater Chem C*, 2015, 3: 596–600
 - 39 Li L, Gu L, Lou Z, *et al.* ZnO quantum dot decorated Zn₂SnO₄ nanowire heterojunction photodetectors with drastic performance enhancement and flexible ultraviolet image sensors. *ACS Nano*, 2017, 11: 4067–4076
 - 40 Lv L, Xu Y, Fang H, *et al.* Generalized colloidal synthesis of high-quality, two-dimensional cesium lead halide perovskite nanosheets and their applications in photodetectors. *Nanoscale*, 2016, 8: 13589–13596
 - 41 Li H, Zhitomirsky D, Grossman JC. Tunable and energetically robust PbS nanoplatelets for optoelectronic applications. *Chem Mater*, 2016, 28: 1888–1896
 - 42 Song J, Xu L, Li J, *et al.* Monolayer and few-layer all-inorganic perovskites as a new family of two-dimensional semiconductors for printable optoelectronic devices. *Adv Mater*, 2016, 28: 4861–4869
 - 43 Lim YR, Song W, Han JK, *et al.* Wafer-scale, homogeneous MoS₂ layers on plastic substrates for flexible visible-light photodetectors. *Adv Mater*, 2016, 28: 5025–5030
 - 44 Dong Y, Gu Y, Zou Y, *et al.* Improving all-inorganic perovskite photodetectors by preferred orientation and plasmonic effect. *Small*, 2016, 12: 5622–5632
 - 45 Dong Y, Zou Y, Song J, *et al.* An all-inkjet-printed flexible UV photodetector. *Nanoscale*, 2017, 9: 8580–8585
 - 46 Vanheusden K, Warren WL, Seager CH, *et al.* Mechanisms behind green photoluminescence in ZnO phosphor powders. *J Appl Phys*, 1996, 79: 7983–7990
 - 47 Lin B, Fu Z, Jia Y. Green luminescent center in undoped zinc oxide films deposited on silicon substrates. *Appl Phys Lett*, 2001, 79: 943–945
 - 48 Kennedy J, Murmu PP, Manikandan E, *et al.* Investigation of structural and photoluminescence properties of gas and metal ions doped zinc oxide single crystals. *J Alloys Compd*, 2014, 616: 614–617
 - 49 Yu D, Cai B, Cao F, *et al.* Cation exchange-induced dimensionality construction: From monolayered to multilayered 2D single crystal halide perovskites. *Adv Mater Interfaces*, 2017, 4: 1700441
 - 50 Liu X, Yu D, Cao F, *et al.* Low-voltage photodetectors with high responsivity based on solution-processed micrometer-scale all-inorganic perovskite nanoplatelets. *Small*, 2017, 13: 1700364
 - 51 Peng M, Wang Y, Shen Q, *et al.* High-performance flexible and broadband photodetectors based on PbS quantum dots/ZnO nanoparticles heterostructure. *Sci China Mater*, 2019, 62: 225–235
 - 52 Peng M, Xie X, Zheng H, *et al.* PbS quantum dots/2D nonlayered CdS_xSe_{1-x} nanosheet hybrid nanostructure for high-performance broadband photodetectors. *ACS Appl Mater Interfaces*, 2018, 10: 43887–43895
 - 53 Alwada'i N, Haque MA, Mitra S, *et al.* High-performance ultraviolet-to-infrared broadband perovskite photodetectors achieved via inter-/intraband transitions. *ACS Appl Mater Interfaces*, 2017, 9: 37832–37838
 - 54 Sun H, Tian W, Cao F, *et al.* Ultrahigh-performance self-powered flexible double-twisted fibrous broadband perovskite photodetector. *Adv Mater*, 2018, 30: 1706986
 - 55 Sahatiya P, Reddy K CS, Badhulika S. Discretely distributed 1D V₂O₅ nanowires over 2D MoS₂ nanoflakes for an enhanced

broadband flexible photodetector covering the ultraviolet to near infrared region. *J Mater Chem C*, 2017, 5: 12728–12736

- 56 Wang P, Liu S, Luo W, *et al.* Arrayed van der Waals broadband detectors for dual-band detection. *Adv Mater*, 2017, 29: 1604439
- 57 Yu J, Chen X, Wang Y, *et al.* A high-performance self-powered broadband photodetector based on a $\text{CH}_3\text{NH}_3\text{PbI}_3$ perovskite/ZnO nanorod array heterostructure. *J Mater Chem C*, 2016, 4: 7302–7308
- 58 Liu H, Zhang X, Zhang L, *et al.* A high-performance photodetector based on an inorganic perovskite–ZnO heterostructure. *J Mater Chem C*, 2017, 5: 6115–6122
- 59 Cao F, Tian W, Gu B, *et al.* High-performance UV–vis photodetectors based on electrospun ZnO nanofiber-solution processed perovskite hybrid structures. *Nano Res*, 2017, 10: 2244–2256
- 60 Gao T, Zhang Q, Chen J, *et al.* Performance-enhancing broadband and flexible photodetectors based on perovskite/ZnO-nanowire hybrid structures. *Adv Opt Mater*, 2017, 5: 1700206

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Author contributions Song J supervised the project; Wang S performed the experiments and wrote the manuscript; Zhu Z and Liu S took part in the fabrication and the characterization of ZnO NWs film; Zou Y and Song J performed the data analysis and revised the manuscript; Dong Y took part in the characterizations of the samples; Xue J and Xu L took part in the preparation of CsPbBr_3 NSs; Dong Y tested the *I*–*V* and *I*–*t* plots of the flexible photodetectors. All authors contributed to the general discussion.

Conflict of interest The authors declare no conflict of interest.

Supplementary information Supplementary results are available in the online version of the paper.

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低维结构策略构筑基于钙钛矿纳米片/氧化锌纳米线的柔性宽光谱探测器

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摘要 柔性光电探测器具有轻便、易携带和优异的大面积兼容性等特点, 在下一代光电子器件领域具有巨大的应用潜力。柔性光电探测器面临的主要挑战是在反复弯曲、拉伸、折叠等形变状态下难以保持优异的性能。本文通过低维度结构策略构筑了基于 CsPbBr_3 纳米片和 ZnO 纳米线的柔性光电探测器。得益于一维纳米线和二维纳米片的高柔性, 所构筑的光电探测器在各种应力下表现出优异的工作稳定性。例如, 在弯曲1000次之后, 器件的性能没有明显变化。此外, 由于 ZnO 和 CsPbBr_3 自身的光吸收特性, 所构筑的柔性光电探测器展现出宽光谱光电响应能力(涵盖紫外和可见波段)。在紫外和可见区域的峰值响应度分别为 3.10 和 0.97 A W^{-1} , 其相应的探测率分别为 5.57×10^{12} 和 $1.71 \times 10^{12} \text{ Jones}$ 。本文针对柔性、高性能集成光电探测器提出的维度构筑策略, 在未来智能、可穿戴光电子器件领域有着巨大的应用前景。